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Sampling Approaches for Measuring Emission Rates from Hazardous Waste Disposal Facilities

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16. ABSTRACT

Fugitive air emission data have been collected for treatment, storage, and disposal operations to support a Regulatory Impact Analysis (RIA) for Hazardous Waste Disposal Facilities. Five sampling approaches have been used for measuring air emission rates from these operations. They are: 1) emission isolation flux chamber, 2) vent sampling, 3) concentration-profile technique, 4) transect technique, and 5) mass balance. Each of these techniques are compared as to their precision, potential applications, and specific limitations of the approaches. Data are presented from sites at which the approaches were applied. Results are compared and contrasted where different techniques are compared as to their precision, potential applications, and specific limitations of the approaches. Data are presented from sites at which the approaches were applied. Results are compared and contrasted where different techniques were used to measure emission rates at the same operation. Finally, recommendations are made for selection of an appropriate sampling approach based on the field experience gained.

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Introduction

The Environmental Protection Agency is in the process of conducting a Regulatory Impact Analysis (RIA) for air emissions from treatment, storage, and disposal operations at hazardous waste disposal facilities. In support of this RIA, data are required which define the levels of air emissions from these facilities. The operations of concern include landfills, surface impoundments, storage tanks, containers (drums), solvent recovery processes and landtreatment. The air emissions from these operations are typically fugitives and as such the operations can be viewed as an area source. The air emissions of primary concern are volatile organic compounds, specifically those compounds or classes of compounds which pose a potential health risk. Sampling approaches for such fugitive emissions have not been standardized in the manner that they have been for ducted emission sources. This paper presents a variety of sampling approaches which have been used to measure the emission rates of volatile organic compounds from four different treatment, storage and disposal facilities (TSDF), discusses their application, limitations, and results which have been obtained from their use at these facilities.

Description of Sampling Approaches

The sampling approaches for measuring air emissions from TSDFs include: 1) emission isolation flux chamber, 2) vent sampling, 3) concentration-profile, 4) transect technique, and 5) mass balance. These approaches can be classified as direct techniques (1 and 2) and indirect techniques (3, 4, and 5). These sampling approaches should be differentiated from the sampling and analytical techniques used to collect and/or analyze the samples. For the field sampling programs described here, air sample collection was by gas-tight syringe (on-site analysis) or evacuated stainless-steel canisters (off-site analysis).[1] Sample analysis was performed on site using a field portable GC-FID (1 ppmv-C detection limit), and off site using a capillary column GC-FID/PID/HECD with cryogenic concentration and sub-ambient temperature programming (1 ppbv-C detection limit).[1] Any liquid or solid samples were collected in glass containers, in a manner which would minimize any headspace. Analysis of the liquid and solid samples were performed using the GC-FID/PID/HECD.

Emission Isolation Flux Chamber

The emission isolation flux chamber is a device used to make a direct emission measurement. The enclosure approach has been used by researchers to measure emission fluxes of sulfur and volatile organic species.[2,3,4] The approach uses an enclosure device (flux chamber) to sample gaseous emissions from a defined surface area. Clean dry sweep air is added to the chamber at a fixed controlled rate. The volumetric flow rate of sweep air through the chamber is recorded and the concentration of the species of interest is measured at the exit of the chamber. The emission rate is expressed as:

$$E_i = C_i R/A \qquad \text{(Equation 1)}$$

where,

E_i = emission rate of component i, ug/m^2-sec
C_i = concentration of component i in the air flowing from the chamber, ug/m^3
R = flow rate of air through the chamber, m^3/sec
A = surface area enclosed by the chamber, m^2

All parameters in Equation 1 are measured directly.

A diagram of the flux chamber apparatus used for measuring emission rates is shown in Figure 1. The sampling equipment consists of a stainless steel/acrylic chamber with impellor, ultra high purity sweep air and rotameter for measuring flow into the chamber, and a sampling manifold for monitoring and/or collection of the specie(s) of interest. Concentrations of total hydrocarbons are monitored continuously in the chamber outlet gas stream using portable flame ionization detector (FID)- and/or photoionization detector (PID)-based analyzers. Samples are collected for subsequent gas chromatographic (GC) analysis once a steady-state emission rate is obtained. Air and soil/liquid temperatures are measured using a thermocouple.

To determine the emission rate for a source of much greater area than that isolated by the flux chamber, a sufficient number of measurements must be taken at different locations to provide statistical confidence limits for the mean emission rate. The area sources measured were gridded and a minimum of six (6) measurements made (when possible) to account for spatial variability. Additionally, a single point was selected as a control point to define temporal variability. On-site GC analyses were performed for all flux chamber measurements and several canister samples were collected for each area to allow off-site detailed GC analysis. Prior to using the chamber, blank and species recovery data were obtained.

Vent Sampling

Methods for measuring emissions from ducted sources are well documented.[5] The approach requires that the volumetric flow rate of the gas be determined, typically as measurements of velocity and duct cross-sectional area, and that the gas concentration be measured. The emission rate can then be calculated as:

$$E_i = C_i U A \qquad \text{(Equation 2)}$$

where,

E_i = emission rate of component i, ug/sec
U = gas velocity through vent, m/sec
C_i = concentration of component i in vent gas, ug/m^3
A = cross-sectional area of vent, m^2

All parameters in Equation 2 are measured directly.

Concentration-Profile

The concentration-profile (C-P) technique was developed by L. J. Thibodeaux and coworkers at the University of Arkansas under a U.S. Environmental Protection Agency contract.[6]. The C-P technique, as developed by Thibodeaux, has been used to estimate emission rates of volatile species from wastewater treatment ponds,[6,7,8] and more recently from landtreatment facilities.[9] The C-P approach is an indirect sampling technique predicated upon experimental measurements of wind velocity, volatile species concentration and temperature profiles in the boundary layer above the waste body. These measurements are used to estimate the vertical flux of the volatile species as:

$$E_i = \left(\frac{D_i}{D_{H_2O}}\right)^n S_v S_i K^2 / \phi_m^2 S_c \qquad \text{(Equation 3)}$$

where,

E_i = emission rates (flux) of organic species i, g/cm^2-sec
D_i = molecular diffusivity of organic species i in air, cm^2/sec
D_{H_2O} = molecular diffusivity of water vapor in air, cm^2/sec
K = von Karman's constant,
S_v = logarithmic slope of the air velocity profile, cm/sec
S_i = logarithmic slope of the concentration-profile for organic species i, g/cm^3
ϕ_m = Businger wind shear parameter
S_c = turbulent Schmidt number
n = exponent for diffusivity ratio

The term $(\phi_m^2 S_c)^{-1}$ represents an atmospheric stability correction factor and is expressed as a function of the Richardson number. The function is an empirical correlation which corrects the estimated emission rate for water vapor to measured values under various atmospheric stabilities. For this reason, the correction factor is valid only under specific meteorological conditions. The molecular diffusivities of water and many organic species are available in the open literature.[10,11] Diffusivities for those compounds for which values are not available, and compound classes or total hydrocarbons must be estimated.

A diagram of the C-P sampling system is shown in Figure 2. The sampling equipment consists of the following: a 4-meter mast with a wind direction indicator, wind speed sensors, temperature sensors, and air collection probes spaced at six logarithmic intervals; a continuous real-time data collection system; a thermocouple for measuring water temperature; and water sampling equipment.

Prior to sample collection, meteorological conditions were monitored for twenty (20) minutes to determine compliance with the necessary

meteorological criteria, a canister blank was taken, and the air collection probes were purged. Once acceptable meteorological conditions were documented, a twenty (20) minute sample collection period was initiated. During the sample collection period, wind speed, air temperature, water temperature, and relative humidity were measured and water samples were obtained. Provisions were made for duplicate air samples to be taken from one of the air collection probes. An upwind air sample was also collected, although it is not expressly required by the methodology. A total of three C-P runs were performed for each of the area sources.

Transect Technique

The transect technique is an indirect emission measurement approach which has been used to measure fugitive particulate and gaseous emissions from area and line sources.[12,13] Horizontal and vertical arrays of samplers are used to measure concentrations of volatile specie(s) within the effective cross-section of the fugitive emission plume. A normal concentration distribution or curve is fitted to the measured concentrations. The volatile specie(s) emission rate is then obtained by spatial integration of the concentrations over this assumed plume area:

$$E_i = uA_s \iint_{A_p} C_{i(h,w)}\, dhdw \qquad \text{(Equation 4)}$$

where,

E_i = emission rate of component i, ug/m^3-sec
u = wind speed, m/sec
C_i = concentration of component i at point (h,w), corrected for upwind background, ug/m^3
h = vertical distance coordinate, m
w = horizontal distance coordinate, m
A_s = surface area of emitting source, m^2
A_p = effective cross-sectional area of plume, m^2

A diagram of the transect sampling system is shown in Figure 3. The sampling equipment consists of a central 3.5 meter mast having three equally spaced air sampling probes and single wind direction, wind speed, and temperature sensors at the top, and five 1.5 meter masts with single air sampling probes. The central mast is aligned with the expected plume centerline. Two masts are placed at equal spacings on each side of the central mast and one mast is used to collect air samples at an upwind location. The spacing of the associated masts are selected to cover the expected horizontal plume cross-section, as defined by oservation and/or profiling with a real-time total hydrocarbon (THC) analyzer.

Prior to sample collection, meteorological parameters were monitored for 20 minutes to determine if acceptable conditions existed, canister blanks were obtained, and the air sampling probes were purged. Following documentation of acceptable meteorological conditions, a 20-minute sampling period was initiated. During the sampling period, meteorological parameters

were monitored. A total of three transect runs were made at each of the area sources.

Mass Balance

Theoretically, emissions or losses from any process can be estimated from an accurate mass balance. If all inlet and outlet process streams are precisely characterized with regard to flow rates, composition, and physical properties, any difference between the total known amount of material entering the system and that known to be leaving would be losses. This can be expressed as:

$$\text{Mass Losses} = \text{Mass In} - \text{Mass Out} \qquad \text{(Equation 5)}$$

In practice, precise measurements of material volumes, flow rates, and characteristics are often difficult to obtain. Most flow rates and material rate measurements in chemical processing are made in terms of volume. Thus, fluid densities must be known to convert volumetric measurements to mass flows. A liquid material balance can be expressed as:

$$E_i = \sum_j L_j W_{i,j} P_j - \sum_k L_k W_{i,k} P_k \qquad \text{(Equation 6)}$$

where,

E_i = emissions (losses) of component i, lb
L_j = volume of inlet stream j, gallons
L_k = volume of outlet stream k, gallons
$W_{i,j}$ = weight fraction of component i in inlet stream j
$W_{i,k}$ = weight fraction of component i in outlet stream k
P_j, P_k = density of liquid stream j and k, respectively, lb/gal

All parameters in Equation 6 are measured. The emissions can also be expressed as a percentage of the total solvent throughput of the process.

Comparison of Methods

The sampling approaches presented are discussed below with regard to their applications/limitations, cost of operation including equipment, manpower and analysis, and the expected precision/sensitivity inherent to the methods.

Applications/Limitations

Table I lists each of the sampling approaches, the TSDF sources for which they can be used to make emission measurements, and limitations which must be considered when using the method. No single sampling approach can be applied to all TSDF sources. When selecting a sampling approach, the specific source must be considered, as well as environmental factors which may limit the ability to make the measurement.

The degree to which environmental factors such as wind speed, temperature, barometric pressure, and relative humidity influence emission rates

from TSDFs is not well defined. Measurement approaches such as the concentration-profile and transect techniques measure emission rates under existing environmental conditions. They are not applicable during periods of low wind speeds (less than 5 mph) or very high wind speeds (greater than 20 mph). The flux chamber technique measures emission rates under virtually no wind speed. The sweep air does provide an air exchange, however, which prevents a concentration buildup at the air interface. The extent to which the emission rate measured with a flux chamber is biased due to isolation from environmental factors has not been determined, but is currently under investigation.

Cost of Operation

The costs considered when comparing the various sampling approaches include equipment costs, manpower and time necessary for the field testing, and off-site analysis. Specific values have been avoided and relative numbers used for the comparison. Table II provides the cost of operation data for each of the sampling approaches. The information provided is based on the experience gained using the various approaches in field applications. The time required to complete a source characterization must consider the time which can be lost when meteorological conditions do not meet the criteria for concentration-profile or transect sampling. Analytical costs reflect the fact that increased sensitivity is necessary for the ambient (more diluted) air samples obtained during transect and C-P sampling, as opposed to the more concentrated samples obtained for the flux chamber, vent sampling, and mass balance sampling approaches.

Precision and Sensitivity

The precision inherent to each of the sampling approaches was assessed using Monte Carlo simulations. Each input parameter for a specific sampling approach was perturbed randomly to simulate the actual variation that would occur in practice. The input parameters were assumed to have a Gaussian distribution, with a standard deviation that was estimated from existing data, prior experience, literature values, and/or actual field measurements. An emission rate was computed from the simulated input of these values and repeated for 100 trials. The calculated variability of each sampling approach is listed in Table III as a coefficient of variation.

These precision estimates are based only on the measurement techniques and do not take into account environmental variations. As such, they represent a best-case value. When using the flux chamber technique, the variation of small areas of emissions ("hot spots") will impact the precision of estimating the overall site emission. The C-P technique will be less affected by "hot spot" emissions, and the transect technique, which is designed to integrate area emissions, should not be significantly affected. However, only the flux chamber technique will be able to detect "hot spot" emissions over a site. The C-P technique and transect technique will be affected by natural, atmospheric fluctuations in wind speed, wind direction, and atmospheric stability. The flux chamber technique will not be affected by

atmospheric fluctuations. The mass balance technique will be influenced to an extreme extent by the ability to obtain homogeneous samples of the process waste.

The lowest detectable emission rate capable of being measured by the sampling approaches can be estimated assuming comparable sample collection and analytical sensitivities. For area sources, the flux chamber has an estimated lower detection level two orders of magnitude lower than the C-P technique and three orders of magnitude lower than the transect technique. This difference explains why the flux chamber can use less sensitive analytical techniques and still obtain comparable detection levels to the C-P and transect techniques.

Results of Field Measurements

The sampling approaches discussed above were used to measure emission rates of volatile organic compounds from four hazardous waste disposal facilities. The area sources tested included active and inactive landfills, landtreatment, surface impoundments, drum storage, tanks, and solvent recovery. Results of the measurements are presented in Table IV and discussed below according to sampling approach. For simplicity, only total nonmethane hydrocarbon emission rates are presented. Where different sampling approaches were used to measure emissions from the same source, the results were compared to determine if there was a significant difference between the reported values. This was accomplished using a statistical test (z-value) calculated from the values to be compared and their corresponding standard deviations.

The flux chamber approach was simple and straightforward to execute in the field. The statistical sampling approach (gridding) does appear suited to the surface impoundments (5-1, 5-2, and 5-6), landtreatment plot (2-LF) and landfill cell with a homogeneous waste (2-Q). However, the landfills with large cells (5-10 and 4-D) accepting a variety of waste types resulted in highly variable data, making an average emission rate questionable as to its representativeness. The average emission rates for the single cells of landfill 5-10 were, however, consistent with the emission rate for the entire landfill as measured by the transect technique. No emissions were detected at the surfaces of the inactive capped landfills (5-7 and 4-C). The flux chamber resulted in significantly greater emission rates for the surface impoundment (5-6), but lower emission rates for the landtreatment plot (2-LF) when compared to results obtained by the C-P technique. The variability in the emission rates determined using the flux chamber was typically much less than with the transect or C-P techniques. In general, very good correlations were observed between all components detected using the chamber and the volatile components in the corresponding waste.

The ability of the vent sampling technique to measure emission rates from vents of inactive landfills (5-7 and 4-C) and storage tanks (6) suffered due to the intermittent and very low gas flow rates encountered. While it was possible to measure concentrations of volatile species in the

vents, the inability to measure a flow rate prevented calculation of an emission rate. Future field tests will use the flux chamber sealed over the vents to measure emission rates. The vent sampling technique is sound for situations where gas flow rates are constant and readily measured, such as the ventilated drum storage building (5-DS).

The C-P technique provided valid data for some compounds but not others when compared to the volatile components in the waste at both the surface impoundment (5-6) and landtreatment plot (2-LF). Logarithmic fits to wind speed and temperature were in most cases valid. The difficulty in obtaining a valid logarithmic concentration-profile in many cases must be attributed to the low concentrations of components and associated variability at such low detection levels. The variability associated with the emission rates measured by the C-P technique was typically less than with the transect technique.

The transect technique was the only sampling approach used which was appropriate for making emission rate measurements during placement of waste at the active landfill (5-10). Concentrations of some components did approximate a normal distribution across the sampling array, allowing the plume area to be estimated and a concentration flux calculated. The plume area was much better defined during active placement of the waste. Transect measurements at active landfill 2-Q resulted in highly variable component concentrations across the sampling array, as did measurements from surface impoundment 6-3. Valid definition of the plume cross-section for both of these sources was questionable. For all applications of the transect technique, low component concentrations, upwind backgrounds, and variability in plume location/concentrations contributed to the difficulty in obtaining a valid plume definition.

The mass balance technique resulted in measurable emission rates from the solvent recovery operation (6-TCE and 6-MEK). In both cases, however, the variability in the measured emission rate was quite large. Improved precision could be obtained by better volumetric measurement techniques.

Conclusions

When selecting a sampling approach, it is desirable to have an accurate and a precise technique to allow comparison of data. Comparable data will best be obtained by using techniques with low inherent variability (e.g., the flux chamber). This is preferred even if a small, known bias (e.g., isolation from environmental factors) exists in the technique. In the case of the sampling approaches discussed above, the accuracy of the techniques has not been determined. However, experience with the sampling approaches has provided information on the variability of the techniques.

Based upon the information gained, the following statements can be made concerning the use of the various sampling approaches. When possible, the emission isolation flux chamber should be used to make measurements of emission rates from waste bodies due to its inherently greater sensitivity

and lower variability. The flux chamber is not well suited to measuring emissions from extremely large waste bodies of varied composition (large spatial variability) or highly agitated liquid surfaces. For waste bodies where the flux chamber is not appropriate, the C-P technique is preferred over the transect technique. A decision to use one or the other technique must, however, be based on the TSDF source, environmental factors, etc. When using the vent sampling technique, consideration must be given to the ability to make the necessary velocity (volumetric flow rate) measurement. The mass balance approach suffers from the fact that the emissions (losses) are typically small, and thus the difference between two large values. The resulting emission rate will typically have a large variability. Whenever possible, the mass balance approach should not be used as the sole means for determining an emission rate from a TSDF source.

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Table I. Application of emission rate measurement sampling approaches to TSDF emission sources.

Sampling Approach	TSDF Source	Limitations/ Comments
Emission Isolation Flux Chamber	active landfills,	small cells with uniform waste composition
	inactive landfills,	can be used on surface and for vents at inactive landfills
	surface impoundments,	little or no surface agitation
	landtreatment	
Vent Sampling	vented treatment systems, vented landfills, vented storage buildings, storage tanks	intermittent/low flow makes flow measurements difficult
Concentration-Profile	surface impoundments, landtreatment	meteorological conditions must meet criteria; not suited for small impoundments or landtreatment plots
Transect Technique	active landfills, surface impoundments, landtreatment, drum storage	meteorological conditions must meet criteria; requires minimal interferences from other upwind sources
Mass Balance	solvent recovery,	must identify and be capable of measuring all streams
	surface impoundment, landtreatment	must assume degradation

Table II. Relative costs of making emission rate measurements at TSDFs using various sampling approaches.

Sampling Approach	Relative Costs				
	Equipment[a]	Manpower[b]	Time[d]	Analytical[h]	Total[i]
Emission Isolation Flux Chamber	5	2	1	1	8
Vent Sampling	5	2	>1[e]	1	>8
Concentration-Profile	10	3[c]	>1[f]	10	>23
Transect Technique	7	2	>1[f]	10	>19
Mass Balance	1	1	>1[g]	1	>3

[a] equipment and time to prepare for sampling

[b] minimum manpower necessary on site for sampling and analysis

[c] assumes 2 men on boat, 1 on shore for safety

[d] days to complete source characterization on site

[e] intermittent emissions can extend time required

[f] inappropriate meteorological conditions can extend time required

[g] time selected based on process

[h] based on number and type of analysis required

[i] equipment + (manpower x time) + analytical

Table III. Estimates of variance for emission rates measured using various sampling approaches.

Sampling Approach	Coefficient of Variation
Emission Isolation Flux Chamber	33%
Vent Sampling	36%
Concentration-Profile	52%
Transect Technique	110%
Mass Balance	34%

Table IV. Emission rates of total nonmethane hydrocarbons from TSDF sources measured using various sampling approaches

TSDF Source	Sampling Approach	Emission Rate (Kg-C/hectare-day)
Active Landfill		
5-10	Transect Technique	3.8, 9.2[a]
	Emission Isolation Flux Chamber	4.5[b], 13[b]
4-D	Emission Isolation Flux Chamber	4.1[b]
2-Q	Emission Isolation Flux Chamber	0.8[b]
Inactive Landfill		
5-7	Emission Isolation Flux Chamber	<0.1[c]
	Vent Sampling	<0.01 Kg-C/day
4-C	Emission Isolation Flux Chamber	<0.1[c]
	Vent Sampling	<0.001 Kg-C/day
Surface Impoundments		
5-1	Emission Isolation Flux Chamber	10
5-2	Emission Isolation Flux Chamber	49
5-6	Emission Isolation Flux Chamber	2.7
	Concentration-Profile	0.8
6-3	Transect Technique	54[d]
Solvent Recovery		
6-TCE	Mass Balance	1.2%[e], 16.7%[f]
6-MEK	Mass Balance	1.1%[e]
Landtreatment		
2-LF	Emission Isolation Flux Chamber	626-53[g]
	Concentration-Profile	1080-831[h]
Drum Storage Building		
5-DS	Vent Sampling	0.2 Kg-C/day
Storage Tanks		
6	Vent Sampling	<0.1 Kg-C/day[i]

[a] active dumping of waste

[b] single cell of landfill

[c] below detection limit

[d] validity of data questionable

[e] distillation losses only

[f] washing losses

[g] emission rates measured from time of spreading to two days after spreading

[h] emission rates measured from one to two days after spreading

[i] no detectable gas flow rate

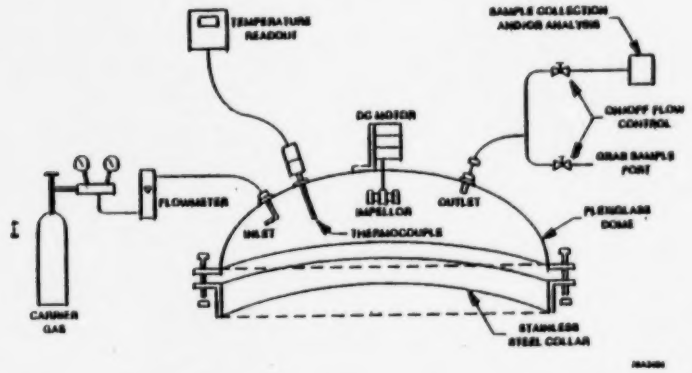

Figure 1. Cutaway side view of emission isolation flux chamber and sampling apparatus.

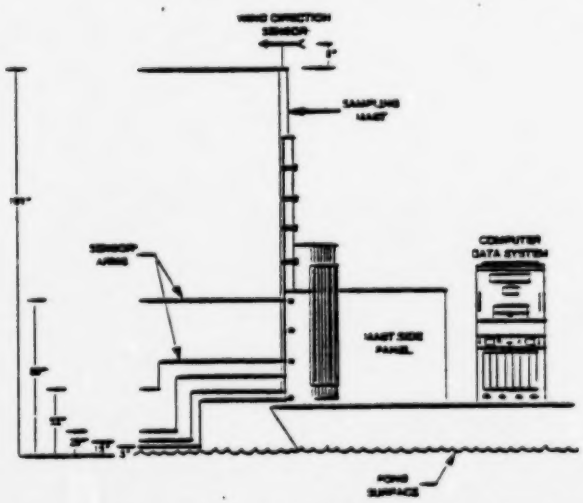

Figure 2. Mast sample collection system for C-P sampling.

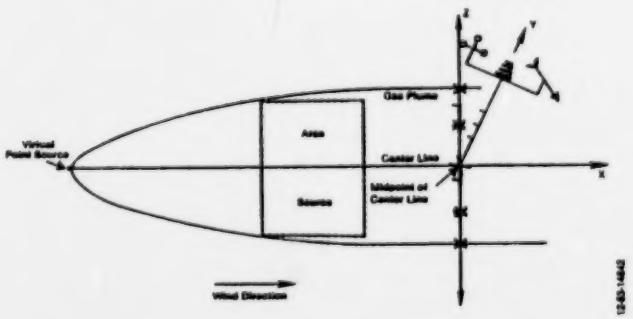

Figure 3. Example of transect technique sampling.